

**TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371**

ATTORNEY DOCKET NUMBER IN-12095	U.S. APPLICATION NO. (SEE 37 CFR 1.53) 10/019998	
INTERNATIONAL APPLICATION NO. PCT/EP 00/003229	INTERNATIONAL FILING DATE 11.04.00	PRIORITY DATE CLAIMED 20.04.99

PREPARATION OF POLYURETHANES

Reinhard LORENZ; Stephan BAUER; Dieter JUNGE; Eva BAUM; Kathrin HARRE; Jörg ERBES; Thomas OSTROWSKI; Georg GROSCH.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371 (b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(C)(2)).
7. ☒ Amendment to the claims of the International Application under PCT Article 19 (35 U.S.C.371(c)(3))
 - a. ☒ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annex to the International Preliminary Examination Report under PCT Article 36

Items 11. to 16. below concern other document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included
13. ☒ A FIRST preliminary amendment.
- ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A Change of power of attorney and/or address letter.
16. ☒ Other items or information: Postcard

**A copy of the cover sheet from the PCT Published Application
Notification of the Recording of a Change (Form PCT/1B/306)**

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No.EL 900586165US addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231

on 10/19/2001

Linda J. Cochran
LINDA J. COCHRAN


U.S. APPLICATION NO. 10/019998 (If known see 37 CFR 1.49)		INTERNATIONAL APPLICATION NO. PCT/EP 00/003229		ATTORNEY'S DOCKET NUMBER IN-12095	
17. <input checked="" type="checkbox"/> The following fees are submitted				CALCULATIONS PTO USE ONLY	
Basic National Fee (37 CFR 1.492(a)(1)-(5)):					
Search Report has been prepared by the EPO or JPO.....				\$890.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482)				\$710.00	
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).....				\$740.00	
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO.....				\$1,040.00	
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4).....				\$100.00	
ENTER APPROPRIATE BASIC FEE AMOUNT				=	\$890.00
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	8 - 20 =	0	X \$18.00	\$0.00	
Independent claims	2 - 03 =	0	X \$84.00	\$0.00	
Multiple dependent claims(s) (if applicable)			+ \$280.00	\$280.00	
TOTAL OF ABOVE CALCULATION				=	\$1170.00
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$	
SUBTOTAL				=	\$1170.00
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				+	\$
TOTAL NATIONAL FEE				=	\$1170.00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				+	\$40.00
TOTAL FEES ENCLOSED				=	\$1210.00
				Amount to be:	\$
				Refunded	
				Charged	\$1210.00

- a. ☐ A check in the amount of \$ _____ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 23-3425 in the amount of \$1210.00 to cover the above fees
A triplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 23-3425. A triplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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Correspondence Information

Correspondence Customer Number:: 28484

Application Information

Title Line One:: PREPARATION OF POLYURETHANES
Application Type:: Utility
Docket Number:: 12095

Representative Information

Representative Customer Number:: 28484

Prior Foreign Applications

Foreign Application One::	19917897.6
Filing Date::	04-20-99
Country::	Germany
Priority Claimed::	Yes

10/019998

531 Rec'd PCT/PTO 19 OCT 2001

PATENT

(Docket No. IN-12095)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

REINHARD LORENZ
ET AL

Serial No.: NEW

Filed: HERewith

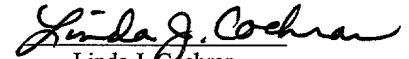
For: PREPARATION OF
POLYURETHANES

Group Art Unit: NEW

Examiner: NEW

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Linda J. Cochran

PRELIMINARY AMENDMENT

BOX PCT APPLICATION

Assistant Commissioner of Patents
Washington, D.C. 20231

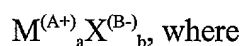
Sir:

In reference to the above-referenced patent application, please enter the following amendment and consider the accompanying remarks prior to examination thereof on the merits.

IN THE CLAIMS:

Please amend the claims as follows:

1. (Amended) A process for producing polyurethanes comprising reacting at least one polyisocyanate with at least one compound containing at least two hydrogen atoms which are reactive toward isocyanate groups, wherein the compound containing at least two active hydrogen atoms comprises at least one polyether alcohol prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide catalysis and wherein the reaction is carried out in the presence of at least one metal salt of the formula



M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , and Ba^{2+} ,

X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- , IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- ,

CO_3^{2-} , S^{2-} , SH^- , HSO_3^- , SO_3^{2-} , HSO_4^- , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$, $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$,

$H_2PO_4^{2-}$, PO_4^{3-} , $P_2O_7^{4-}$, $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_2)^-$, $(S_{n+1}H_{sn-2}O_4)^{2-}$ where $n = 1-20$ and

their mixed salts and mixtures,

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

with the proviso that the compound is electrically neutral.

2. (Amended) A process as claimed in claim 1, wherein the metal salt $M^{(A+)}_a X^{(B-)}_b$ is selected such that:

$M^{(A+)} = Li^+$, Na^+ , K^+ , NH_4^+ , Mg^{2+} , or Ca^{2+} , and

$X^{(B-)} = F^-$, Cl^- , Br^- , I^- , NO_3^- , HCO_3^- , CO_3^{2-} , HSO_4^- , SO_4^{2-} , $H_2PO_4^-$, HPO_4^{2-} , PO_4^{3-} , $(OC_nH_{2n+1})^-$,

$(C_nH_{2n-1}O_2)^-$, or $(C_{n+1}H_{2n-2}O_4)^{2-}$ where $n = 1-20$

and their mixed salts and mixtures, where

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

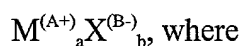
with the proviso that the compound is electrically neutral.

4. (Amended) A process as claimed in claim 1 or 2, wherein the metal salt is dissolved in the polyisocyanate.

6. (Amended) A polyurethane produced according to any one of the processes as claimed in claims 1 to 5.

7. (Amended) A flexible polyurethane foam produced according to any one of the processes as claimed in claims 1 to 6.

8. (Amended) A polyether alcohol comprising the reaction product of H-functional compounds with alkylene oxides using multimetal cyanides as catalysts comprising at least one metal salt of the formula



M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , and Ba^{2+} ,

X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- , IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- ,

CO_3^{2-} , S^{2-} , SH^- , HSO_3^- , SO_3^{2-} , HSO_4^- , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$, $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$,

$H_2PO_4^{2-}$, PO_4^{3-} , $P_2O_7^{4-}$, $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_2)^-$, $(S_{n+1}H_{sn-2}O_4)^{2-}$ where $n = 1-20$ and

their mixed salts and mixtures,

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

with the proviso that the compound is electrically neutral.

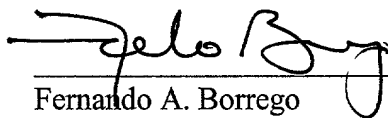
REMARKS

Applicants respectfully request examination of the present application as amended herein. Claims 1, 2, 4, and 6-8 have been amended. Upon entry of the above preliminary amendment, claims 1-8 remain pending in the application. A marked-up version of the amended claims is attached hereto in Appendix A. Should the Examiner have any questions, please contact the undersigned attorney.

Respectfully submitted,

Date:

10/19/01

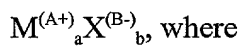


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APPENDIX A

1. (Amended) A process for producing polyurethanes [by] comprising reacting at least one polyisocyanate with at least one compound containing at least two hydrogen atoms which are reactive toward isocyanate groups, wherein the compound containing at least two active hydrogen atoms [which is used is] comprises at least one polyether alcohol prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide catalysis and wherein the reaction is carried out in the presence of at least one metal salt of the formula



M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , and Ba^{2+} ,

X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- , IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- ,

CO_3^{2-} , S^{2-} , SH^- , HSO_3^- , SO_3^{2-} , HSO_4^- , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$, $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$,

$H_2PO_4^{2-}$, PO_4^{3-} , $P_2O_7^{4-}$, $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_2)^-$, $(S_{n+1}H_{sn-2}O_4)^{2-}$ where $n = 1-20$ and

their mixed salts and mixtures, [where]

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

with the proviso that the compound is electrically neutral.

2. (Amended) A process as claimed in claim 1, wherein the metal salt $M^{(A+)}_a X^{(B-)}_b$ is selected [from among] such that:

$M^{(A+)} = Li^+$, Na^+ , K^+ , NH_4^+ , Mg^{2+} , or Ca^{2+} , and

$X^{(B-)} = F^-$, Cl^- , Br^- , I^- , NO_3^- , HCO_3^- , CO_3^{2-} , HSO_4^- , SO_4^{2-} , $H_2PO_4^-$, HPO_4^{2-} , PO_4^{3-} , $(OC_nH_{2n+1})^-$,

$(C_nH_{2n-1}O_2)^-$, or $(C_{n+1}H_{2n-2}O_4)^{2-}$ where $n = 1-20$

and their mixed salts and mixtures, where

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

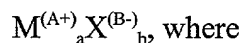
with the proviso that the compound is electrically neutral.

4. (Amended) A process as claimed in [any of claims 1 to 3] claim 1 or 2, wherein the metal salt is dissolved in the polyisocyanate.

6. (Amended) A polyurethane [which can be] produced according to any one of the processes as claimed in [any of] claims 1 to 5.

7. (Amended) A flexible polyurethane foam [which can be] produced according to any one of the processes as claimed in [any of] claims 1 to 6.

8. (Amended) A polyether alcohol [which can be prepared by reacting] comprising the reaction product of H-functional compounds with alkylene oxides using multimetal cyanides as catalysts [and which contains] comprising at least one metal salt of the formula



M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , and Ba^{2+} ,

X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- , IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- ,

CO_3^{2-} , S^{2-} , SH^- , HSO_3^- , SO_3^{2-} , HSO_4^- , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$, $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$,

$H_2PO_4^{2-}$, PO_4^{3-} , $P_2O_7^{4-}$, $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_2)^-$, $(S_{n+1}H_{sn-2}O_4)^{2-}$ where $n = 1-20$ and

their mixed salts and mixtures,

A^+ is the valence of the cation,

B^- is the valence of the anion and

a and b are integers,

with the proviso that the compound is electrically neutral.

Preparation of polyurethanes

Polyurethanes are produced in large quantities. An important
5 starting material used for their production is polyether
alcohols. The latter are usually prepared by catalytic addition
of lower alkylene oxides, in particular ethylene oxide and
propylene oxide onto H-functional initiator molecules. The
catalysts used are usually basic metal hydroxides or salts, with
10 potassium hydroxide having the greatest industrial importance.

In the synthesis of polyether polyols having long chains and
hydroxyl numbers of from about 26 to about 60 mg KOH/g, which are
used for producing flexible polyurethane foams in particular,
15 secondary reactions which lead to faults in the chain structure
occur as chain growth progressors. These by-products are referred
to as unsaturated constituents and lead to impairment of the
properties of the resulting polyurethane materials. In
particular, these unsaturated constituents, which have an
20 OH-functionality of 1, have the following consequences:

- Owing to their sometimes very low molecular weight, they are
volatile and thus increase the total content of volatile
constituents in the polyether polyol and in the polyurethanes
25 produced therefrom, in particular flexible polyurethane
foams.
- They act as chain terminators in the production of the
polyurethane because they retard or reduce the crosslinking
30 of the polyurethane or the buildup of the molecular weight of
the polyurethane.

It is therefore very desirable in industry to avoid unsaturated
constituents as far as possible.

35 One way of preparing polyether alcohols having a low content of
unsaturated constituents is the use of multimetal cyanide
catalysts, usually zinc hexacyanometalates, as alkoxylation
catalysts. There is a large number of documents which describe
40 the preparation of polyether alcohols by means of such catalysts.
Thus, DD-A-203 735 and DD-A-203 734 describe the preparation of
polyether polyols using zinc hexacyanocobaltate. The use of
multimetal cyanide catalysts enables the content of unsaturated
constituents in the polyether polyol to be reduced to about
45 0.003 - 0.009 meq/g; in the case of conventional catalysis using

potassium hydroxide, about 10 times these amounts are found (about 0.03 - 0.08 meq/g).

The preparation of the zinc hexacyanometalates is also known.

- 5 These catalysts are usually prepared by reacting solutions of metal salts such as zinc chloride with solutions of alkali metal or alkaline earth metal cyanometalates, e.g. potassium hexacyanocobaltate. A water-miscible, heteroatom-containing component is generally added to the resulting suspension
- 10 immediately after the precipitation. This component can also previously be present in one or both starting solutions. This water-miscible heteroatom-containing component can be, for example, an ether, polyether, alcohol, ketone or mixture thereof. Such processes are described, for example, in US 3,278,457,
- 15 US 3,278,458, US 3,278,459, US 3,427,256, US 3,427,334, US 3,404,109, US 3,829,505, US 3,941,849, EP 283,148, EP 385,619, EP 654,302, EP 659,798, EP 665,254, EP 743,093, EP 755,716, EP 862,947, EP 892,002, US 4,843,054, US 4,877,906, US 5,158,922, US 5,426,081, US 5,470,813, US 5,482,908, US 5,498,583,
- 20 US 5,523,386, US 5,525,565, US 5,545,601, JP 7,308,583, JP 6,248,068, JP 4,351,632 and US-A-5,545,601.

- One problem with the use of polyether alcohols which have been prepared by means of multimetal cyanide catalysts is their lower
- 25 reactivity compared to other polyether alcohols catalyzed by means of hydroxides. In order to rectify this deficiency, WO 94/03519 proposes adding alkali metal oxides and hydroxides and/or alkaline earth metal oxides and hydroxides in an amount of from 0.5 to 10 ppm to the polyether alcohols prepared by means of
- 30 multimetal cyanide catalysts after removal of the catalyst.

- However, it has been found that polyether alcohols which have been prepared by means of multimetal cyanide catalysts and to which the compounds described in WO 94/03519 have been added
- 35 cannot be used for producing flexible polyurethane foams. The foams produced in this way have severe cracks running through the entire test specimen. In addition, severe dark-brown discoloration of the foam specimens is observed.

- 40 If the addition of metal compounds to polyether alcohols which have been prepared by means of multimetal cyanide catalysts is omitted entirely, their reactivity is, as indicated above, insufficient. Crack formation occurs in many cases. If the amount of catalyst, in particular the amount of tin catalyst, is
- 45 increased, crack formation can be prevented but the foams are usually closed-celled. The foamability of the polyetherols is regarded by the foam manufacturer as being better if the amount

of tin catalysts added to the A component comprising polyols, catalysts, blowing agents and auxiliaries can be varied over as wide as possible a range while at the same time achieving satisfactory open-cellularity, satisfactory mechanical properties
 5 and a homogeneous structure of the foam.

It is an object of the present invention to increase the reactivity of polyether alcohols which have been prepared by means of multimetal cyanide catalysts without the abovementioned
 10 disadvantages being encountered in the production of flexible polyurethane foams.

We have found that this object is achieved by adding metal salts to polyether alcohols which have been prepared by means of
 15 multimetal cyanide catalysts.

The present invention accordingly provides polyether alcohols which can be prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide
 20 catalysts and to which metal salts are added.

The invention further provides a process for producing polyurethanes, in particular flexible polyurethane foams, by reacting isocyanates with compounds containing at least two
 25 active hydrogen atoms, wherein the compounds containing at least two active hydrogen atoms which are used are polyetherols which can be prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide catalysts and to which metal salts are added.

30 The invention also provides polyurethanes, in particular flexible polyurethane foams, which can be prepared by reacting isocyanates with compounds containing at least two active hydrogen atoms, wherein the compounds containing at least two active hydrogen
 35 atoms which are used are polyetherols which can be prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide catalysts and the reaction is carried out in the presence of metal salts.

40 The metal salts used are preferably ones of the formula
 $M^{(A+)}_a X^{(B-)}_b$ where

M is an alkali metal or alkaline earth metal, an ammonium ion, boron, aluminum or hydrogen and

45 X is an organic or inorganic anion, where

4

A^+ is the valence of the cation,
 B^- is the valence of the anion and
 a and b are integers,

5 with the proviso that the compound is electrically neutral.

Preference is given to using metal salts of the formula
 $M^{(A+)}_a X^{(B-)}_b$ in which

- 10 $M^{(A+)} =$ $Li^+, Na^+, K^+, Rb^+, Cs^+, NH_4^+, Be^{2+}, Mg^{2+}, Ca^{2+}, Sr^{2+}, Ba^{2+}$
 and
 $X^{(B-)} =$ $F^-, Cl^-, ClO^-, ClO_3^-, ClO_4^-, Br^-, I^-, IO_3^-, CN^-,$
 $OCN^-, NO_2^-, NO_3^-, HCO_3^-, CO_3^{2-}, S^{2-}, SH^-, HSO_3^-, SO_3^{2-}, HSO_4^-,$
 $SO_4^{2-}, S_2O_2^{2-}, S_2O_3^{2-}, S_2O_4^{2-}, S_2O_5^{2-}, S_2O_6^{2-}, S_2O_7^{2-}, S_2O_8^{2-},$
 15 $H_2PO_2^-, H_2PO_4^-, HPO_4^{2-}, PO_4^{3-}, P_2O_7^{4-}, (OC_nH_{2n+1})^-,$
 $(C_nH_{2n-1}O_2)^-, (C_{n+1}H_{2n-2}O_4)^{2-}$
 where $n = 1-20$

and their mixed salts and mixtures, where

- 20 A^+ is the valence of the cation,
 B^- is the valence of the anion and
 a and b are integers,

25 with the proviso that the compound is electrically neutral.

Particular preference is given to salts of the formula $M^{(A+)}_a X^{(B-)}_b$
 in which

- 30 $M^{(A+)} =$ $Li^+, Na^+, K^+, NH_4^+, Mg^{2+}, Ca^{2+},$ and
 $X^{(B-)} =$ $F^-, Cl^-, Br^-, I^-, NO_3^-, HCO_3^-, CO_3^{2-}, HSO_4^-, SO_4^{2-}, H_2PO_4^-,$
 $HPO_4^{2-}, PO_4^{3-}, (OC_nH_{2n+1})^-, (C_nH_{2n-1}O_2)^-, (C_{n+1}H_{2n-2}O_4)^{2-}$
 where $n = 1-20$

35 and their mixed salts and mixtures, where

- A^+ is the valence of the cation,
 B^- is the valence of the anion and
 40 a and b are integers,

with the proviso that the compound is electrically neutral.

Particular preference is given to potassium salts, in particular
 45 potassium chloride, potassium phosphate and potassium acetate.

The salts are preferably used in an amount of from 0.5 to 50 ppm, based on the A component. The salts are preferably added to the polyether alcohols and the addition is preferably carried out after the preparation of the polyether alcohols, before or in particular after the purification step for the polyether alcohols. However, it is also possible to add the salts prior to the polyether alcohol synthesis, preferably by addition to the initiator substance. The salts are inert in the preparation of the polyether alcohols. In order to obtain optimum mixing of the salts with the polyether alcohol, they are preferably added as solution. Water can be used as solvent, but it is also possible to first mix the salts with part of the polyether alcohol and then add this mixture to the polyether alcohol.

In a specific embodiment of the invention, the salts can be added to a blowing agent, in particular water. In a further embodiment of the invention, the salts can also be added to a catalyst and/or an auxiliary. In a further embodiment of the invention, the salts can also be added to the isocyanate component.

According to the present invention, the metal salts can also be added to more than one component.

The preparation of the polyether alcohols is carried out, as indicated above, by catalytic addition of alkylene oxides, in particular ethylene oxide, propylene oxide, butylene oxide, vinylloxirane, styrene oxide or mixtures of at least two of the abovementioned alkylene oxides. The use of the catalysts according to the present invention is particularly advantageous in the addition reaction of propylene oxide, ethylene oxide and their mixtures.

Initiator substances used are, as indicated above, H-functional compounds. For preparing the polyether alcohols of the present invention, preference is given to using bifunctional and/or trifunctional alcohols, preferably ethylene glycol, diethylene glycol, propylene glycol, dipropylene glycol, 1,4-butanediol, glycerol, trimethylolpropane, pentaerythritol, water or any mixtures of at least two of the compounds mentioned.

The multimetal cyanide compounds used as catalysts have the formula



where

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- M¹ is a metal ion selected from the group consisting of Zn²⁺, Fe²⁺, Co³⁺, Ni²⁺, Mn²⁺, Co²⁺, Sn²⁺, Pb²⁺, Mo⁴⁺, Mo⁶⁺, Al³⁺, V⁴⁺, V⁵⁺, Sr²⁺, W⁴⁺, W⁶⁺, Cr²⁺, Cr³⁺, Cd²⁺, Hg²⁺, Pd²⁺, Pt²⁺, V²⁺, Mg²⁺, Ca²⁺, Ba²⁺, Cu²⁺ and mixtures thereof,
- 5 M² is a metal ion selected from the group consisting of Fe²⁺, Fe³⁺, Co²⁺, Co³⁺, Mn²⁺, Mn³⁺, V⁴⁺, V⁵⁺, Cr²⁺, Cr³⁺, Rh³⁺, Ru²⁺, Ir³⁺ and M¹ and M² are identical or different,
- 10 A is an anion selected from the group consisting of halide, hydroxide, sulfate, carbonate, cyanide, thiocyanate, isocyanate, cyanate, carboxylate, oxalate and nitrate,
- L is a water-miscible ligand selected from the group consisting of alcohols, aldehydes, ketones, ethers, polyethers, esters, ureas, amides, nitriles and sulfides,
- 15 b, c and d are selected so that the compound is electrically neutral and
- 20 e is the number of coordinated ligands and,
- f is the number of coordinated water molecules.
- 25 The catalysts used can be X-ray-amorphous, as described in EP-A-654,302 or EP-A-743,093. The catalysts can also be X-ray-crystalline with an X-ray diffraction pattern as described in EP-A-755,716.
- 30 The catalysts can be prepared either from the alkali metal or alkaline earth metal cyanometallates as described in EP-A-555,053 or from the cyanometalic acids as described in EP-A-862,947.

The catalysts can be used as powder or as pastes. The use of
35 pastes is described in WO 97/26,080.

It is also possible to use catalysts in which the multimetal cyanide compounds are applied to or introduced into inert organic and/or inorganic supports or shaped to form catalyst-containing
40 shaped bodies.

The molecular addition of the alkylene oxides is carried out under the customary conditions at from 60 to 180°C, preferably from 90 to 140°C, in particular from 100 to 130°C, and pressures
45 in the range from 0 to 15 bar, preferably in the range from 0 to 10 bar and in particular in the range from 0 to 5 bar. The initiator substance/DMC mixture can be pretreated by stripping

before commencement of the alkoxylation, as taught in WO 98/52,689.

- After the molecular addition of the alkylene oxides is complete,
- 5 the polyether alcohol is worked up according to customary methods by removing the unreacted alkylene oxides and volatile constituents, usually by distillation, steam stripping or gas stripping and/or other methods of deodorization. If necessary, a filtration can also be carried out.
- 10 The catalyst can be deactivated and/or removed from the polyol. Usually, the catalyst is, if desired with the help of filter aids, filtered off and/or centrifuged off.
- 15 As indicated above, the polyether alcohols of the present invention are preferably reacted with polyisocyanates to produce polyurethanes, preferably polyurethane foams and thermoplastic polyurethanes, in particular flexible polyurethane foams. In this reaction, the polyether alcohols of the present invention can be
- 20 used individually or in admixture with other compounds containing at least two active hydrogen atoms.

- Polyisocyanates used here can be any isocyanates having two or more isocyanate groups in the molecule. It is possible to use
- 25 either aliphatic isocyanates such as hexamethylenediisocyanate (HDI) or isophorone diisocyanate (IPDI), or preferably aromatic isocyanates, such as tolylene diisocyanate (TDI), diphenylmethane diisocyanate (MDI) or mixtures of diphenylmethane diisocyanate and polyphenylpolymethylene polyisocyanates (crude MDI). It is
- 30 also possible to use isocyanates which have been modified by incorporation of urethane, uretdione, isocyanurate, allophanate, uretonimine and other groups, known as modified isocyanates.

- As compounds containing at least two groups which are reactive
- 35 toward isocyanate groups, which are used in admixture with the polyether alcohols of the present invention, it is possible to use amines and mercaptans, but preferably polyols. Among the polyols, polyether polyols and polyester polyols have the greatest industrial importance. The polyether polyols used for
- 40 producing polyurethanes are usually prepared by base-catalyzed addition of alkylene oxides, in particular ethylene oxide and/or propylene oxide, onto H-functional initiator substances. Polyester polyols are usually prepared by esterification of polyfunctional carboxylic acids with polyfunctional alcohols.

The compounds containing at least two groups which are reactive toward isocyanates also include chain extenders and/or crosslinkers which can also be used if desired. These are at least bifunctional amines and/or alcohols having molecular weights in the range from 60 to 400.

- As blowing agents, use is usually made of water and compounds which are gaseous at the temperature of the urethane reaction and are inert toward the starting materials for the polyurethanes, known as physically active blowing agents, and also mixtures thereof. Physically acting blowing agents used are hydrocarbons having from 2 to 6 carbon atoms, halogenated hydrocarbons having from 2 to 6 carbon atoms, ketones, acetals, ethers and inert gases such as carbon dioxide or noble gases.
- Catalysts used are, in particular, amine compounds and/or metal compounds, in particular heavy metal salts and/or organic metal compounds. Particular preference is given to using known tertiary amines and/or organic metal compounds as catalysts. Suitable organic metal compounds are, for example, tin compounds such as tin(II) salts of organic carboxylic acids, e.g. tin(II) acetate, tin(II) octoate, tin(II) ethylhexanoate and tin(II) laurate, and the dialkyl tin(IV) salts of organic carboxylic acids, e.g. dibutyltin diacetate, dibutyltin dilaurate, dibutyltin maleate and dioctyltin diacetate. Examples of organic amines which are customary for this purpose are: triethylamine, 1,4-diazabicyclo[2.2.2]octane, tributylamine, dimethylbenzylamine, N,N,N',N'-tetramethylethylenediamine, N,N,N',N'-tetramethylbutanediamine, N,N,N',N'-tetramethylhexane-1,6-diamine, dimethylcyclohexylamine, pentamethyldipropylenetriamine, pentamethyldiethylenetriamine, 3-methyl-6-dimethylamino-3-azapentol, dimethylaminopropylamine, 1,3-bis(dimethylamino)butane, bis(2-dimethylaminoethyl) ether, N-ethylmorpholine, N-methylmorpholine, N-cyclohexylmorpholine, 2-dimethylaminoethoxy ethanol, dimethylethanolamine, tetramethylhexamethylenediamine, dimethylamino-N-methylethanolamine, N-methylimidazole, N-formyl-N,N'-dimethylbutylenediamine, N-dimethylaminoethylmorpholine, 3,3'-bis(dimethylamino)di-n-propylamine and/or bis(2-piperazinoisopropyl) ether, diazabicyclo[2.2.2]octane, dimethylpiperazine, N,N'-bis(3-aminopropyl)ethylenediamine and/or tris(N,N-dimethylaminopropyl)-s-hexahydrotriazine, 4-chloro-2,5-dimethyl-1-(N-methylaminoethyl)-imidazole, 2-aminopropyl-4,5-dimethoxy-1-methylimidazole, 1-aminopropyl-2,4,5-tributylimidazole, 1-aminoethyl-4-hexylimidazole, 1-aminobutyl-2,5-dimethylimidazole,

- 1-(3-aminopropyl)-2-ethyl-4-methylimidazole,
1-(3-aminopropyl)imidazole and/or
1-(3-aminopropyl)-2-methylimidazole, preferably
1,4-diazabicyclo[2.2.2]octane and/or imidazoles, particularly
5 preferably 1-(3-aminopropyl)imidazole,
1-(3-aminopropyl)-2-methylimidazole and/or
1,4-diazabicyclo[2.2.2]octane. The catalyst described can be used
individually or in the form of mixtures.
- 10 Auxiliaries and/or additives used are, for example, mold release
agents, flame retardants, colorants, fillers and/or reinforcing
materials.
- It is customary in industry to combine all starting materials
15 with the exception of the polyisocyanates to form a polyol
component and to react this with the polyisocyanates to give the
polyurethanes.
- The polyurethanes can be produced by the one-shot method or by
20 the prepolymer method. The flexible polyurethane foams can be
either slabstock foams or molded foams.
- An overview of the starting materials for producing polyurethanes
and the processes employed for this purpose may be found, for
25 example, in Kunststoffhandbuch, Volume 7 "Polyurethane",
Carl-Hanser-Verlag Munich Vienna, 1st Edition 1966, 2nd Edition
1983 and 3rd Edition 1993.
- Surprisingly, the flexible foams produced in the presence of
30 salts from the DMC-catalyzed polyether alcohols display a
significantly improved curing behavior, without crack formation,
compared to those to which no additives have been added.
- Polyether alcohols to which hydroxides have been added as per the
35 process described in WO 94/03519 cannot be processed to give
flexible polyurethane foams. The foam produced in this way has
severe cracks running through the entire test specimen. In
addition, severe dark-brown discoloration occurs.
- 40 In contrast, the polyether alcohols of the present invention can
be processed to give flexible polyurethane foams having a high
open-cellularity or high air permeability and a defect-free foam
structure without crack formation.
- 45 The invention is illustrated by the examples below.

Example 1

Preparation of the catalyst

- 5 7 l of strong acid ion exchanger in the sodium form (Amberlite 252 Na, Rohm&Haas) are introduced into an ion exchange column (length 1 m, volume 7.7 l). The ion exchanger is subsequently converted into the H-form by passing 10% strength hydrochloric acid through the ion exchange column for 9 hours at a flow rate
- 10 of 2 bed volumes per hour until the Na content of the eluate is less than 1 ppm. The ion exchanger is subsequently washed with water until neutral.

- The regenerated ion exchanger is then used to prepare an
- 15 essentially alkali-free hexacyanocobaltic acid ($H_3[Co(CN)_6]$). For this purpose, a 0.24 M solution of potassium hexacyanocobaltate ($K_3[Co(CN)_6]$) in water is passed over the ion exchanger at a flow rate of 1 bed volume per hour. After 2.5 bed volumes, the potassium hexacyanocobaltate solution is replaced by water. The
- 20 2.5 bed volumes obtained have, on average, a hexacyanocobaltic acid content of 4.5% by weight and an alkali metal content of less than 1 ppm.

- 2000 ml of an aqueous hexacyanocobaltic acid solution (4.4% by
- 25 weight of $H_3[Co(CN)_6]$, K content < 1 ppm) are heated to 40°C and subsequently admixed while stirring (blade stirrer, 500 rpm) with a solution of 178.8 g of zinc(II) acetate dihydrate in 600 g of water. 350 g of tert-butanol are subsequently added to the suspension. The suspension is stirred at 40°C for another
- 30 30 minutes. The solid is then filtered off with suction and washed on the filter with 2000 ml of tert-butanol. The solid which had been treated in this way was dried under reduced pressure at 50°C for 16 hours.
- 35 The X-ray diffraction pattern of the double metal cyanide obtained in this way indicated a monoclinic structure.

Example 2

- 40 Preparation of the polyether alcohol

- In the following example, the initiator substance used was a oligopropylene-glycerol having a hydroxyl number of 370 mg KOH/g which had been obtained by an alkali-catalyzed reaction of
- 45 glycerol with propylene oxide at 105°C. The hydroxyl number was determined in accordance with ASTM D 2849.

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- 605 g of the oligopropylene-glycerol were mixed with 2.14 g of the catalyst from Example 1 under a nitrogen atmosphere in a stirring autoclave. After evacuation of the autoclave, a mixture of 20 g of ethylene oxide and 130 g of propylene oxide was
- 5 metered in at 125°C. The almost immediate commencement of the reaction was recognized by an only brief pressure rise to 2.5 bar which was followed by an immediate pressure drop. After 10 minutes no free propylene oxide or ethylene oxide remained in the reactor. A mixture of 565.5 g of ethylene oxide and 3690 g of
- 10 propylene oxide was then fed in at the same temperature at such a rate that a pressure of 2.6 bar abs. was not exceeded. The metering-in phase was complete after only 100 minutes and after a further 20 minutes the reaction phase was complete, as could be seen from the pressure signal.
- 15 The polyetherol obtained in this way was filtered once using a deep-bed filter. The polyol had a hydroxyl number of 48 mg KOH/g, a viscosity at 25°C of 632 mPa•s, a content of unsaturated constituents of 0.0044 meq/g, a zinc content of 23 ppm and a
- 20 cobalt content of 11 ppm.

Examples 3-7

Production of the polyisocyanate polyaddition products

- 25 Polyol A: Polyether alcohol as described in Example 2
- Polyol B: 1 g of a 33% strength aqueous potassium hydroxide solution was mixed with 100 g of polyol A.
- 30 Polyol C: 1 g of a 33% strength aqueous potassium phosphate solution was mixed with 100 g of polyol A.
- Lupragen® N201: 1,4-diazabicyclo[2.2.2]octane (33%) in
- 35 dipropylene glycol (67%), (BASF Aktiengesellschaft)
- Lupragen® N206: bis-(2-dimethylaminoethyl) ether (70%) in
- 40 dipropylene glycol (30%), (BASF Aktiengesellschaft)
- Kosmus® 29: Tin(II) salt of ethylhexanoic acid, (Goldschmidt AG)
- 45 Tegostab® BF 2370: Silicone stabilizer (Goldschmidt AG)

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Lupranat® T80: 2,4-/2,6-tolylene diisocyanate mixture
(BASF Aktiengesellschaft)

The abovementioned starting materials were reacted in the ratios
5 indicated in Table 1.

All components apart from the isocyanate Lupranat® T80 A were
first combined by intensive mixing to form a polyol component.
The Lupranat® T80 A was then added while stirring and the
10 reaction mixture was poured into an open mold in which it foamed
to give a polyurethane foam. The data for the foams obtained are
shown in Table 2.

Table 1

15	Example	3	4	5	6
	Polyol A [g]	1000	999	999	999
	Polyol B [g]	-	1	-	-
	Polyol C [g]	-	-	1	
	BF 2370 [g]	10	10	10	10
20	N201/206 3:1 [g]	2	2	2	2
	K 29 [g]	3.4	3.4	3.4	3.4
	Water [g]	45	45	45	45
	T 80 [g]	487.46	487.46	487.46	487.46
	Index	110			

25 Table 2

	Example	3	4	5	6
	Cream time [s]	11	-	10	12
	Fiber time [s]	75	-	80	90
	Rise time [s]	80	-	85	95
30	Rise height [mm]	285	-	285	270
	Rise height after 5 min [mm]	270	-	280	265
	Air permeability [mmWS]	400	-	350	25
35	Appearance of the foam	Crack formation	Unusable foam	Homoge- neous, fine- celled	Homoge- neous, fine- celled

40

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We claim:

1. A process for producing polyurethanes by reacting at least one polyisocyanate with at least one compound containing at least two hydrogen atoms which are reactive toward isocyanate groups, wherein the compound containing at least two active hydrogen atoms which is used is at least one polyether alcohol prepared by addition of alkylene oxides onto H-functional initiator substances by means of multimetal cyanide catalysis and the reaction is carried out in the presence of at least one metal salt of the formula

$M^{(A+)}_a X^{(B-)}_b$, where

M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , Ba^{2+} ,

X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- , IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- , CO_3^{2-} , S^{2-} , SH^- , HSO_3^- , SO_3^{2-} , HSO_4^- , SO_4^{2-} , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$, $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$, $H_2PO_4^-$, HPO_4^{2-} , PO_4^{3-} , $P_2O_7^{4-}$, $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_4)^{2-}$ where $n = 1-20$ and their mixed salts and mixtures, where

A^+ is the valence of the cation,
 B^- is the valence of the anion and
 a and b are integers,

with the proviso that the compound is electrically neutral.

2. A process as claimed in claim 1, wherein the metal salt $M^{(A+)}_a X^{(B-)}_b$ is selected from among:

$M^{(A+)} = Li^+$, Na^+ , K^+ , NH_4^+ , Mg^{2+} , Ca^{2+} , and

$X^{(B-)} = F^-$, Cl^- , Br^- , I^- , NO_3^- , HCO_3^- , CO_3^{2-} , HSO_4^- , SO_4^{2-} , $H_2PO_4^-$, HPO_4^{2-} , PO_4^{3-} , $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_4)^{2-}$ where $n = 1-20$

and their mixed salts and mixtures, where

A^+ is the valence of the cation,
 B^- is the valence of the anion and
 a and b are integers,

with the proviso that the compound is electrically neutral.

3. A process as claimed in claim 1 or 2, wherein the metal salt is dissolved in the compound having at least two active hydrogen atoms.
- 5 4. A process as claimed in any of claims 1 to 3, wherein the metal salt is dissolved in the polyisocyanate.
5. A process as claimed in any of claims 1 to 4, wherein the metal salt is used in an amount of from 0.1 to 50 ppm, based
10 on the compound having at least two active hydrogen atoms.
6. A polyurethane which can be produced as claimed in any of claims 1 to 5.
- 15 7. A flexible polyurethane foam which can be produced as claimed in any of claims 1 to 6.
8. A polyether alcohol which can be prepared by reacting H-functional compounds with alkylene oxides using multimetal
20 cyanides as catalysts and which contains at least one metal salt of the formula
- $M(A^+)_a X(B^-)_b$, where
- 25 M is selected from among Li^+ , Na^+ , K^+ , Rb^+ , Cs^+ , Be^{2+} , Mg^{2+} , Ca^{2+} , Sr^{2+} , Ba^{2+} ,
- X is selected from among F^- , Cl^- , ClO^- , ClO_3^- , ClO_4^- , Br^- , I^- ,
30 IO_3^- , CN^- , OCN^- , NO_2^- , NO_3^- , HCO_3^- , CO_3^{2-} , S^{2-} , SH^- , HSO_3^- ,
 SO_3^{2-} , HSO_4^- , SO_4^{2-} , $S_2O_2^{2-}$, $S_2O_3^{2-}$, $S_2O_4^{2-}$, $S_2O_5^{2-}$, $S_2O_6^{2-}$,
 $S_2O_7^{2-}$, $S_2O_8^{2-}$, $H_2PO_2^-$, $H_2PO_4^-$, HPO_4^{2-} , PO_4^{3-} , $P_2O_7^{4-}$,
 $(OC_nH_{2n+1})^-$, $(C_nH_{2n-1}O_2)^-$, $(C_{n+1}H_{2n-2}O_4)^{2-}$ where $n = 1-20$
and their mixed salts and mixtures, where
- 35 A^+ is the valence of the cation,
 B^- is the valence of the anion and
a and b are integers,
- with the proviso that the compound is electrically neutral.
- 40
- 45

0050/049917

DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that:

INVENTORSHIP IDENTIFICATION

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Preparation of polyurethans

SPECIFICATION IDENTIFICATION

the specification of which

☐ is attached hereto.

☐ was filed on _____ as

Application Serial No. _____

and was amended on _____ (if applicable).

☒ was filed as PCT international application

Number PCT/EP00/03229

on 11/04/2000

and was amended under PCT Article 19

on _____ (if applicable)

ACKNOWLEDGMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information known by me to be material to the patentability of this application in accordance with Title 37, Code of the Federal Regulations. §1.56(a).

☐ In compliance with this duty there is attached an information disclosure statement. 37 CFR 1.97.

☒ In compliance with this duty, information which may be material is disclosed in the specification of the subject application.

CLAIM FOR BENEFIT OF EARLIER U. S. / PCT APPLICATION(S) UNDER 35 U. S. C. 120

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s) or PCT international application(s) designating the United States of America that is / are listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in that / those prior application(s) in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, §1.56(a) which occurred between the filing date of the prior application(s) and the national of PCT international filing date of this application.

U. S. Application(s) (or PCT applications designating U. S.)

U. S. Application Serial No.	Filing Date	Status (pending, patented, abandoned)

RELATED FOREIGN APPLICATIONS

Related foreign applications, if any, filed in the name of the inventor(s) or the inventor(s) assigns more than twelve months before the filing of the subject application are as follows

Country	Application No.	Date of filing	Date of issue or publication

POWER OF ATTORNEY

I hereby appoint the following attorney(s) and agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected herewith:

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 Reg. No. 34,584;
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 Reg. No. 31,278;
 Reg. No. 42,024;
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CLAIM FOR BENEFIT OF FOREIGN PRIORITY UNDER 35 U. S. C. §119

Thereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United State of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

☐ No such applications have been filed.

☒ Such applications have been filed as follows

**DETAILS OF FOREIGN APPLICATION FROM WHICH PRIORITY CLAIMED
UNDER 35 U. S. C. §119**

Country	Application No.	Date of filing	Date of issue or publication
Germany	19917897.6	20 April 1999	

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